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TITLE: A NOVEL 15.9-MICRON SOURCE

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A novel 15.9-micron source

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Abstract

Laser emission has been generated in $^{15}ND_3$ by pumping at 860.4 cm^{-1}. Strong laser action has been observed at 123 cm^{-1}, 109 cm^{-1} and, with up to 10 millijoules extracted, at 628.1 cm^{-1}. Spectroscopic analysis indicates that the pumping arises from, and the 628.1-cm^{-1} emission terminates on, the same rotational state. Analysis of the time histories of the three laser emissions as well as studies of laser output energies indicates that 15.9-micron output arises from a 4-wave type process. This system, in addition, clarifies the interpretation of earlier studies of CO_2-laser-pumped ammonia lasers.

Introduction

Laser isotope separation of uranium hexafluoride led to the development of many new laser systems. Completely tunable sources at 15.9 μm all suffered from the limitation of low pulse power. CO_2-laser-pumped molecular lasers promised high pulse powers as well as high efficiency, but depended on an accidental coincidence of a CO_2 laser line and a molecular absorption connected to a useful 15.9-μm transition. In a previous paper[1] we described an optical pumping experiment that gave an output which we tentatively described as near-resonant parametric conversion.

We report in this paper on the production of 15.9-μm emission from $^{13}CO_2$-laser-pumped $^{15}ND_3$. This system differs from previously reported CO_2-laser-pumped ammonia lasers in ways unrelated to the absorption frequency shifts due to isotopic substitution. The $^{13}CO_2$ laser operates on the P(27) line of the "hot band" $01^11 \rightarrow [03^10, 11^10]_1$ at 860.42 cm^{-1}. The 15.9-μm emission of $^{15}ND_3$ terminates on the same rotational level that the $^{13}CO_2$ laser absorption arises from, making a population inversion unlikely. In the process of increasing the 15.9-μm conversion we made many observations and measurements, which we report in this paper.

Experimental

The pump laser was a Lumonix 103 TEA laser with one Brewster angle NaCl window on the output side and an internal Littrow diffraction grating (PTR master, 90 lines/mm). The output coupler was a coated Ge mirror with a 20-m internal radius and a reflectivity of 90% at the lasing wavelength. A closed-cycle circulation system was operated to force the laser mixture through a heated catalyst[2] chosen to accelerate the oxidation of ^{13}CO produced by the TEA discharge to minimize the steady-state concentration of molecular oxygen. The laser gas composition was optimized at 700 torr to minimize leakage or entry of the local atmosphere (atmospheric pressure in Los Alamos is approximately 580 torr). The usual mixture was 10% $^{13}CO_2$, 10% N_2 and 80% He. ^{13}CO was added in amounts that did not influence laser performance (less than 10 torr) but failed to increase the gas-fill lifetime. The laser output was approximately 1 J at a repetition rate of 0.25 Hz. A modification of the $^{13}CO_2$ laser was the in-cavity insertion[3] of a pulsed low-pressure $^{13}CO_2$ gain cell to force the laser to operate on a single longitudinal mode.

The first experiments were conducted with apparatus shown in Fig. 2 of Ref. 1. The 5-cm bore stainless-steel cell was 1 m in length. The cell ends were spherical, copper mirrors, each having a centered, 2-mm-diam hole, one of which allowed the pump beam entrance. This cell was used for spectroscopy of the deutero-ammonia laser emission, both ir and fir. For fir spectroscopy the output hole was sealed with either a polyethylene or z-cut quartz window. Both ir and fir spectroscopic searches were performed with a 0.5-m monochromator with liquid-N_2-cooled photoconductive Hg-Cd-Te detection in the ir and room-temperature pyroelectric detection in the fir. Higher resolution measurements were made near 15.9 μm with a 1-m Spex monochromator.

Figure 1 illustrates the configuration of an experiment designed to increase the 15.9-μm energy conversion efficiency. The multimode $^{13}CO_2$ output was controlled by an internal iris and the beam was propagated through the center of the cell. Conversion to 15.9 μm was measured by dispersing the cell throughput with a diffraction grating.

The $^{15}ND_3$ was analyzed using mass spectroscopy to be better than 95% pure. The cell and gas-handling system were prepared by repeated fillings and pumping of D_2O to minimize hydrogen-deuterium exchange on windows and walls.

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Results and observations

Absorption of the $^{13}CO_2$ laser radiation in $^{15}ND_3$ varied from 1.3×10^{-2} cm^{-1} $torr^{-1}$ for small signals to 2×10^{-3} cm^{-1} $torr^{-1}$ at a fluence of 1.0 J/cm^2. The pulse length was ~500 ns and showed the structure of passive mode locking. The pumping frequency corresponded to the aR(11,K) band[4] and ir emission was detected immediately in a hole-coupled[1] cavity at a pressure of 1 torr. The observed lack of sensitivity of the output energy to mirror tuning was characteristic of a high-gain ir laser such as CO_2-laser-pumped CF_4.

With a hole-coupled cell, the emission detected in the ir was near 628.1 cm^{-1}. $^{15}ND_3$ pressure was varied over a large range and N_2 or He was added to alter the conditions of collisional transfer. Added gases did not change the output spectrum but only lowered the output energy. At a pressure of 4 torr of pure $^{15}ND_3$, weak emission was measured at 615 cm^{-1} corresponding to the aP(12,K) transitions, but the intensity was always less than 1% of the 628.1-cm^{-1} output. Fir signals were detected at wavelengths of 123 cm^{-1} and 109 cm^{-1} with relative intensities of approximately 10 to 1. High-speed measurement of the structure of the output of the 628.1-cm^{-1} emission or the combined fir output showed the temporal structure characteristic of the mode-locked $^{13}CO_2$ laser input. Figure 2 illustrates the simultaneous $^{13}CO_2$ pump pulse and 628.1-cm^{-1} output and shows the 17-ns periodicity.

To check for waveguide effects, a 1.2-cm bore polished brass tube 1 m in length was inserted into the 5-cm bore stainless-steel tube. Under otherwise unchanged conditions, no ir output was observed. Smoothing of the $^{13}CO_2$ laser led to a smooth 628.1-cm^{-1} output. In the windowed cell configuration (Fig. 1), cell dimensions were varied over a wide range. Lengths of copper cells varied between 0.5 and 6.0 m. The largest ir output of nearly 10 mJ was obtained with the 6.0-m cell, which had a 10-cm bore. With shorter cells, the substitution of pyrex for copper lowered the 628.1-cm^{-1} output by approximately 10%. The output was very sensitive to the focal position of the pump beam. For greatest efficiency, the pump beam diverged throughout the length of the cell. The divergence angle was also important, but a careful study of the angle dependence was not attempted. The output frequencies were all polarized parallel to the pump laser polarization and, in the case of the windowed cell, the 15.9-μm output propagated nearly entirely in the direction of the pump laser. A large number of 15.9-μm spectra were taken in this configuration, an example of which is given in Fig. 3. The experimental configuration was that of Fig. 1, with KCl windows and a pressure of 2.5 torr $^{15}ND_3$. An alteration of pressure, cell length, or focusing configuration changed the detailed shape of the spectral output. Changing the pressure to 3.5 torr, for example, shifted the peak of the output spectrum to coincide with K = 4.

Changing window material allowed a test of the effect of feedback on 628.1-cm^{-1} output. Figure 4 illustrates the results of these experiments. Listed in parentheses are the reflectances due to the Restrahlen effect on the 123-cm^{-1} and the 109-cm^{-1} fir emissions. The CO_2 pump energy was normalized to account for the Fresnel loss in entering the cell. While experiments varying the 628.1-cm^{-1} feedback in grating configurations were inconclusive, single-pass extraction appeared to control the output level.

Replacing $^{15}ND_3$ with $^{14}ND_3$ and tuning the $^{13}CO_2$ laser accordingly led to two new laser lines: The 11P33 and 11P12 lines produced ir emission at 641 cm^{-1} and 619 cm^{-1}, in the same pattern as the $^{15}ND_3$ output.

Discussion

The calculated spectrum[4] of $^{15}ND_3$ has the $^{13}CO_2$ laser center frequency falling between the aR(11,4) and aR(11,5) lines at 860.41 cm^{-1} and 860.42 cm^{-1}, respectively. Doppler broadening in room-temperature $^{15}ND_3$ leads to absorption by lines from aR(11,1) through aR(11,6), without considering power broadening or the spectral width of the pump laser. Figure 3, illustrating the output spectrum and comparing the calculated aP(11,K) spectrum, indicates a similar range of K values in the output. The fine structure could not be resolved in our fir spectra.

Without considering fine structure we characterize the observed processes as a closed cycle. Absorption of the pump radiation by the aR(11,K) band leads to near-resonant Raman stimulated emission at 123 cm^{-1}. Population is transferred to symmetric $v_2 = 1$, $J = 11$ [s(1,11)], with some population remaining in a(1,12). The s(1,11) is inverted with respect to a(1,10), leading to stimulated emission at 109 cm^{-1}. The emission at 628.1 cm^{-1} corresponds to the energy difference between a(1,10) and s(0,11), an allowed ir transition.

Figure 5 is a simplified energy level diagram with the relevant states in the conversion of the 860.4-cm^{-1} pump input to the 628.1-cm^{-1} output. All the observed wavelengths [including the weak aP(12,K) emission at 615 cm^{-1}, not shown] are radiatively coupled; i.e., all transitions are allowed and no radiation was observed which could arise only from collisional transfer. Further evidence against the involvement of collisional processes comes from our observation that the structure in the ir and 628.2-cm^{-1} pulses mirrors the mode-locked pattern in the pump pulse. In addition, peak conversion in this laser occurred at low pressures (< 4 torr) and addition of a buffer gas always reduced conversion.

Emission at 628.1 cm^{-1} cannot be easily characterized by a process requiring population inversion. We have observed all the lasing wavelengths shown in Fig. 5 simultaneously with the hole-coupled cell. In addition, we have seen no evidence of transparency or gain in the $^{13}CO_2$ laser beam which passes through the $^{15}ND_3$ cell. We therefore have the inequality s(0,11) > s(1,11) > a(1,10) referring to the population in those levels (ignoring fine structure and degeneracies). By this inequality it is impossible to have the population inversion a(1,10) > s(0,11), which would lead to stimulated emission at 628.1 cm^{-1}. A properly phased set of short pulses as seen in Fig. 2 would allow all these inversions, but when the $^{13}CO_2$ laser emission was smoothed with an internal gain cell,[3] the 628.1-cm^{-1} signal was also smooth and was generated at the same conversion efficiency as the mode-locked case.

Parametric conversion of the pump and the two simultaneous stimulated fir emission processes seem to be the likely source of 628.1-cm^{-1} emission. Resonances increase the efficiencies of 4-wave processes. Analysis of triply resonant mixing has been performed[5] where two of the three frequencies are nearly the same. The authors point out that resonant absorption has several complicating effects on conversion. They state that "it broadens the phase matching peak, reduces the effective length of interaction and makes the deduction of the dispersion of χ^3...less straightforward." The fine structure in $^{15}ND_3$ makes the spectrum complex and the time-dependent populations change the phase-matching conditions. These complicating factors have made it difficult to understand the interaction of the adjustable experimental parameters.

Conclusion

Observation of simultaneous collisionless emission in the ir and fir occurring in an optically pumped, 4-wave cycle suggests that the 628.1-cm^{-1} emission is parametric rather than the result of a population inversion. Further analysis of this system or similar systems seems necessary to understand this mechanism of shifting a pump laser radiation to longer wavelengths.

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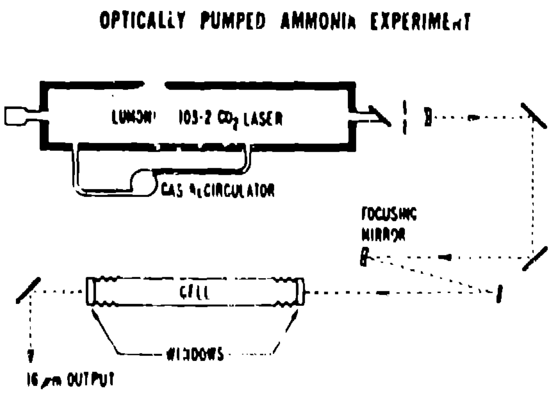

Figure 1. Schematic drawing of the $^{15}ND_3$ laser configuration with minimum feedback used for optimization of 628.1-cm^{-1} conversion.

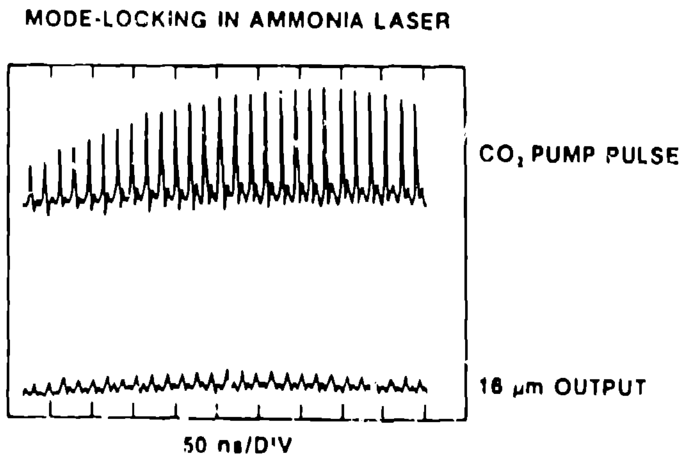

Figure 2. Signals from a pyroelectric detector (Molectron P5-01). Signals were measured simultaneously but the onset of the 15.9-micron emission was obscured by the detector noise.

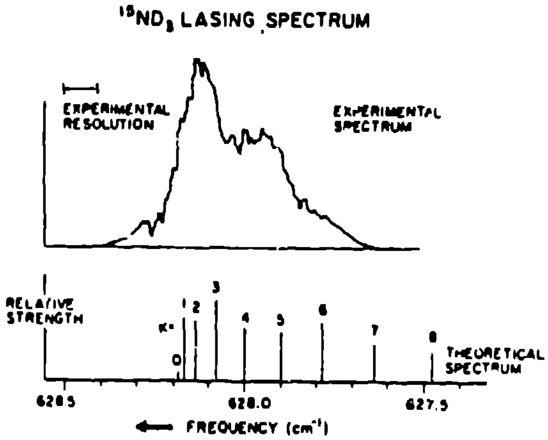

Figure 3. Spectrum of 15.9-μm emission obtained with a 1-m focal-length Spex monochromator. $^{15}ND_3$ pressure was 2.0 torr in a 2-m cell in the configuration of Fig. 1.

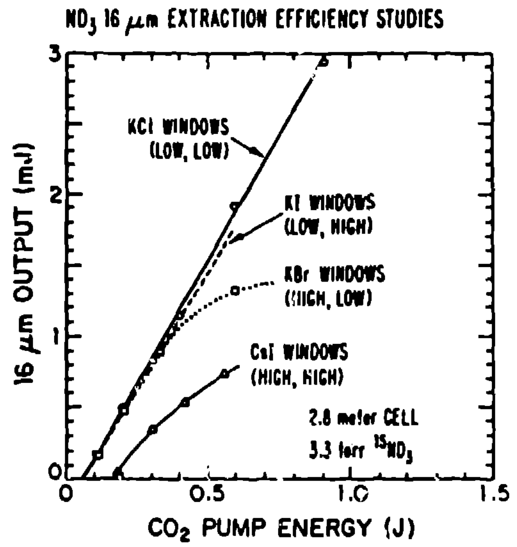

Figure 4. Results of a parametric study of 15.9-μm output. CO_2 pump energy is normalized by the entrance window reflectivity. In brackets are the relative reflectivities of the 123-cm^{-1} and the 109-cm^{-1} emissions, respectively.

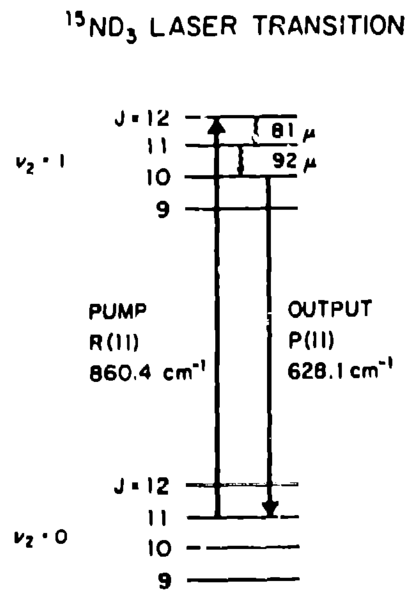

Figure 5. Simplified diagram of the coupled $^{15}ND_3$ levels. Absent from this diagram are the splittings due to the double minimum potential of ν_2 (a and s) and the angular momentum orientation (K < J).